Exact Thermodynamics of Disordered Impurities in Quantum Spin Chains

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Exact results for the thermodynamic properties of ensembles of magnetic impurities with randomly distributed host-impurity couplings in the quantum antiferromagnetic Heisenberg model are presented. Exact calculations are done for arbitrary values of temperature and external magnetic field. We have shown that for strong disorder the quenching of the impurity moments is absent. For weak disorder the screening persists, but with the critical non-Fermi-liquid behaviors of the magnetic susceptibility and specific heat. A comparison with the disordered Kondo effect experiments in dirty metallic alloys is performed.

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In the last few years the interest in non-Fermi-liquid (NFL) behavior of magnetic metals and metallic alloys has grown considerably. A large class of conducting nonmagnetic materials does not behave as usual Fermi liquids (FL) at low temperatures. One of the best known examples of such a behavior is the Kondo effect for multi-(n) channel electron systems: For an impurity spin less than $\frac{n}{2}$ a NFL critical behavior results [1]. The critical behavior of a single magnetic impurity can also be connected with a quadrupolar Kondo effect or non-magnetic two- channel Kondo effect [1]. However, for most of the dirty metals and alloys, in which the NFL behavior was observed, see e.g. the recent review [2] and Refs. [3,4], the magnetic susceptibility (χ) and low temperature specific heat (C) usually show logarithmic or very weak power law behavior with temperature (T). The resistivity linearly decreases with temperature showing a large residual resistivity. The last property together with the alloy nature of the compounds suggests that the disorder (a random distribution of localized f-electrons or a random coupling to the conducting electron host) may play the main role in the low temperature NFL character of such systems. The idea of (non-screened) local moments existing in disordered metallic systems has been already pointed out recently [5–7]. It was proposed that near the metal-insulator transitions (or for the sufficiently alloyed systems far from the quantum critical point) disordered correlated metals contain localized moments. The random distribution of their magnetic characteristics (i.e. their Kondo temperatures, T_K, which are characteristic energy scales for the crossover between the screened, or strong-coupling regime, and the weak coupling behavior) may be connected either with the randomness of exchange couplings of itinerant electrons with the local moments [6], or with the randomness of the densities of conduction electron states [5]. In Ref. [4] the results of the measurements of the magnetic susceptibility, NMR Knight shift and low temperature specific heat have been reported. To explain the features of the behavior it was necessary to assume weak disorder, with a Gaussian distribution of the Kondo temperatures. However the model, which was used for the explanation of the experiment, was oversimplified [4]: The magnetization of a single magnetic moment was approximated by the Brillouin function $B(ah/T + bT_K)$, where h is the external magnetic field, and a, b are constants. It was mentioned in [4] that the data for the specific heat and Knight shift did not agree with the ones predicted by this simple theory, especially for nonzero values of the magnetic field. The disagreement could be caused either by an inadequate representation of the Kondo magnetization by the single-impurity theory, or by the simple replacement $T \to T + bT_K$ in the Brillouin function, or because of the not perfectly symmetric Gaussian distribution of the impurity couplings. The inhomogeneous magnetic susceptibility was confirmed very recently in [8] by muon spin rotation experiments. The role of the long-range (RKKY) coupling between the local moments was taken into account recently in [9] (Griffiths phase theory). The latter gives a qualitatively similar behavior as for models with non-interacting local moments [8].

It is known that the behavior of a single magnetic impurity in a one-dimensional (1D) antiferromagnetic (AF) Heisenberg spin $S = \frac{1}{2}$ chain as well as the behavior of a single Kondo impurity in a 3D free electron host are described by similar Bethe ansatz theories [1,10,11], e.g. the magnetization and the low-temperature magnetic specific heat of the impurity for both models coincide. The spin-$\frac{1}{2}$ Heisenberg model is the seminal model for correlated many-body systems. Most of its static properties are exactly known. The spin-$\frac{1}{2}$ magnetic impurity manifests the total Kondo screening with FL-like low temperature behavior of the magnetic susceptibility and specific heat. In other words, the moment of the impurity is quenched by the host spins, like the one by the spin degrees of freedom of conduction electrons for the Kondo effect. On the other hand, for the integrable lattice models one can incorporate a finite concentration of magnetic impurities [12] without destroying the exact solvability (it is impossible in the free electron host [1], where a single magnetic impurity can only be embedded). Hence, for the random distribution of magnetic impurities we can

suppose that low dimensionality is not principal for the Kondo screening. The absence of the magnetic ordering in the NFL Kondo systems [2] also confirms this assumption and leads to the goal of our present investigation: To find exactly the thermodynamics of the disordered ensemble of spin-$\frac{1}{2}$ magnetic impurities in the Heisenberg chain (with various *random distributions of the impurity-host couplings*) for *arbitrary* values of the external magnetic field and temperature. It is the first study in which the thermodynamic characteristics of a *disordered interacting* many-body system are calculated *exactly* without any approximations. In this Letter we show that: (i) For several kinds of strong disorder of the impurity-host couplings the (Kondo) screening is absent; (ii) for weaker disorder the quenching persists, but with a NFL behavior of the magnetic characteristics. We performed a comparison of our exact results with previous approximate ones and with experimental data of NFL alloys.

We investigate the thermodynamics of the quantum spin-$\frac{1}{2}$ AF chain with spin-$\frac{1}{2}$ impurities. The Hamiltonian of the system has the form $H = \sum_j H_{j,j+1} + H_{imp}$, where the host part is $H_{j,j+1} = \vec{S}_j \vec{S}_{j+1}$ (the host exchange constant J is equated to unity). The impurities' part of the Hamiltonian has the standard form for the exactly solvable lattice Hamiltonians. Suppose we have an impurity distribution in which impurities are not nearest neighbors, then for the impurity, say situated between sites m and $m+1$ of the host we obtain [11,13,14]

$$\begin{aligned} H_{imp} = J_{imp}\big(&H_{m,imp} + H_{imp,m+1} - H_{m,m+1} \\ &+ i\theta[H_{m,imp}, H_{imp,m+1}]\big) \ , \end{aligned} \tag{1}$$

where $J_{imp} = (\theta^2+1)^{-1}$, and $[.,.]$ denotes the commutator. The coupling of the impurity to the host (J_{imp}) is determined by the constant θ. It was shown in [11,13] that precisely this constant determines the effective Kondo temperature of the impurity via $T_K \propto \exp(-\pi|\theta|)$: For energies higher than this crossover Kondo scale one has the asymptotically free impurity spin $\frac{1}{2}$, while for the lower energies the impurity spin is screened, and the usual FL-like behavior persists. In other words, θ measures the shift of the Kondo resonance of the impurity level with the host spin excitations, similar to the standard picture of the Kondo effect in the electron host. We can independently incorporate any number of such impurities into the host chain, each of them will be characterized by its own θ_j. Hence we obtain an ensemble of the spin-$\frac{1}{2}$ impurities with their own Kondo temperatures. The lattice Hamiltonian Eq. (1) has additional terms, which renormalize the coupling between the neighboring sites of the host, and three- spin terms. However it was shown in [13] that in the long-wave limit such a lattice form of the impurity Hamiltonian yields the well-known form of the contact impurity-host interaction similar to the one of the usual Kondo problem [1]. The contact impurity coupling in this (conformal) limit is also determined by the same constant θ.

We managed to map our quantum Hamiltonian at finite temperature to a classical system in 2D by means of a Trotter-Suzuki decomposition [15]. The geometry of this classical system is a square lattice with width L (=length of the quantum chain) and height N (=Trotter number). The interactions on the lattice are four-spin interactions around faces with coupling parameters depending on $(NT)^{-1}$ and the interaction parameter θ_i where i is the number of the column to which the considered face of the lattice belongs to. Note that the interactions are homogeneous in each column, but vary from column to column. We study this system in the limit $N, L \to \infty$ using an approach which is based on a transfer matrix describing transfer in horizontal direction. The corresponding column-to-column transfer matrices are referred to as quantum transfer matrices (QTM). See Fig. 1 for an illustration of the model.

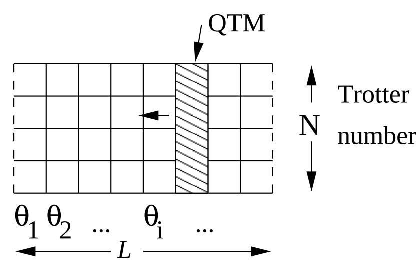

FIG. 1. Illustration of the geometry underlying the classical model with four-spin interaction around faces and alternating coupling parameters from column to column.

In general all QTMs corresponding to the L many columns are different. However, all these operators can be proven to commute pairwise. Therefore, the free energy per lattice site of our system can be calculated from just the largest eigenvalues of the quantum transfer matrices (corresponding to just one eigenstate). For a discussion of the homogeneous case see [17,16] and references therein. For our system we find the following set of non-linear integral equations for "energy density" functions of spinons $a(x)$, $\bar{a}(x)$, $A = 1 + a$ and $\bar{A} = 1 + \bar{a}$ (x is the spectral parameter):

$$\begin{aligned} &\int \big[k(x-y)\ln A(y) - k(x-y-i\pi+i\epsilon)\ln \bar{A}(y)\big]dy \\ &= \ln a(x) - \frac{h}{2T} + \frac{\pi}{T\cosh x} \ , \end{aligned} \tag{2}$$

with kernel function $k(x) = \frac{1}{2\pi}\int d\omega e^{-\pi|\omega|+i2x\omega}/\cosh \pi\omega$. The corresponding equation for $\bar{a}(x)$ is obtained from Eq. (2) by exchanging $i \to -i$, $h \to -h$ and $a, A \leftrightarrow \bar{a}, \bar{A}$. The free energy per site f is given by

$$f(x) = e_0(x) - \frac{T}{2\pi}\int \frac{\ln[A(y)\bar{A}(y)]dy}{\cosh(x-y)} \ , \tag{3}$$

where e_0 is the groundstate energy. The free energy of the total chain with impurities is $F = \sum_j f(\frac{\pi}{2}\theta_j)$, where the sum is taken over all the sites (for sites without impurities we get $f(0)$). These equations are easily solved numerically for arbitrary magnetic field values and temperatures. The random distribution of the values θ_j (or of the Kondo temperatures for the impurities) can be described by a distribution function $P(\theta_j)$. The details of calculations as well as the generalization of our model for the magnetically anisotropic case (important for systems with strong spin-orbital couplings [9]) will be reported later. It is worthwhile to emphasize here the simplicity of the derived equations: For each impurity there is only one parameter, the shift of the spectral parameter in the formula for the free energy per site Eq. (3). Then the exact solvability of the problem for any number of impurities permits to introduce the distribution of these shifts (or the strengths of the impurity-host couplings, i.e. the local Kondo temperatures). One has only two (non-linear) integral equations, Eqs. (2), to solve, and the answer can be obtained for arbitrary temperature (in principle down to $T = 10^{-24}J$) and magnetic field ranges.

In Fig. 2 the results for (a) the magnetic susceptibility, and (b) the linear-temperature coefficient of the specific heat $\gamma = C/T$ at zero field for a homogeneous Heisenberg chain, a single Kondo impurity, a Gaussian distribution of the host-impurity couplings, a Lorentzian distribution, and a so-called logarithmically normal [18] distribution, which is characteristic for strong disorder, e.g. close to a critical point [6]) are plotted as functions of the temperature. One can observe a clear qualitative difference between strong disorder (Lorentz and log-normal) and the other curves. It is clear that the divergent value of the magnetic susceptibility at low T for the strong disorder of the impurity-host couplings is connected with the fact that for most of the impurities their Kondo temperatures are lower than the temperature of the system. Therefore these impurities give rise to a Curie-like behavior of the susceptibility. This divergence disappears upon applying a finite external field which restors most of the FL-like behavior.

Our results for the low-temperature thermodynamics are close to the ones of the perturbative calculations of random AF spin-$\frac{1}{2}$ chains [19,20]: γ and χ have weak power-law or logarithmic singularities. Our low temperature results confirm the very weak dependence of the critical exponents on the temperature. For the log-normal distribution we find critical exponents of 0.134 and 0.131 for $\chi(T)$ and $\gamma(T)$. In the case of the Lorentzian distribution these exponents are 0.846 and 0.730. The rather large deviations of these exponents are due to strong logarithmic corrections in $\chi(T)$ at low temperatures. For an illustration of this effect see also Fig. 3 showing the Wilson ratios γ/χ which are non-universal, i.e. with NFL behavior and show infinite slope at $T = 0$.

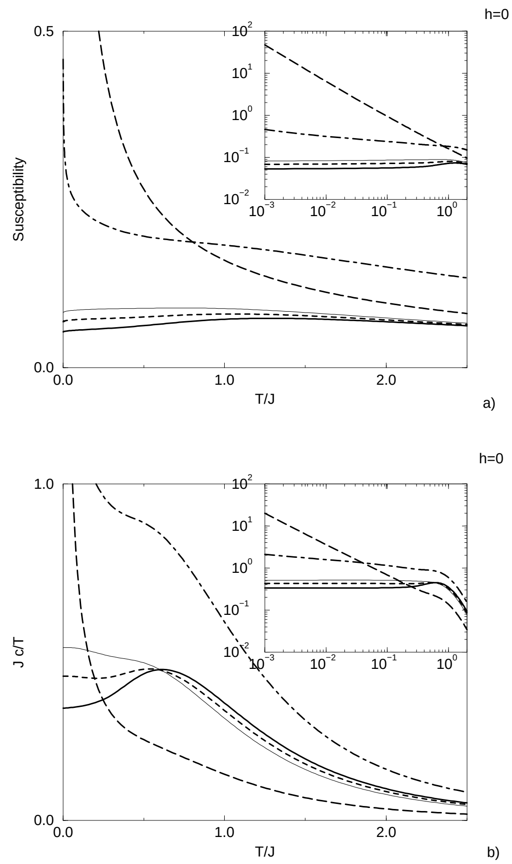

FIG. 2. The temperature dependence of (a) χ and (b) γ at zero magnetic field for: The homogeneous Heisenberg chain (solid line); a single impurity (thin line); Gaussian distribution of the parameter θ (dashed line); Lorentzian distribution (long-dashed line); a log-normal distribution (dashed-dotted line). The insets show log-log plots of the data.

We like to note that former calculations were valid at most for low temperatures, while our approach is applicable for *any* temperature and magnetic field scales. Furthermore, it is also known that the approximate results [19,20] incorrectly give zero or infinite susceptibility at $T \to 0$ irrespective of the distribution, while its true value is finite for e.g. the homogeneous chain and the single impurity [11,16]. Our scheme, of course, perfectly describes the correct behavior. We also performed a comparison of our results with the data of Ref. [4], with qualitatively similar results. Note that any distribution $P(\lambda)$ of impurity couplings $\lambda(\propto 1/\theta$ in our parametrization) with finite $P(0)$ corresponds to a distribution in θ with Lorentzian tail. Hence the agreement of our data with [4,7]: infinite χ and γ for $T \to 0$. More clearly the comparision with experiments can be seen from Refs. [21,22]. By changing the concentration of impurities one can go from weak to strong disorder with quantative agreement with wide and narrow Gaussian and log-normal distributions [21]. Critical exponents very close to those of our

Lorentzian distribution were observed in [22].

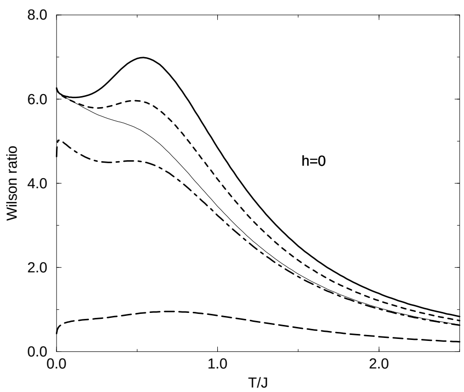

FIG. 3. The temperature dependence of the Wilson ratio [1] at zero magnetic field. Depiction of distributions by lines as in Fig. 2. Note the infinite slope at $T = 0$ due to logarithmic corrections.

Generalizing model (1) by keeping exact solvability, it is possible to include (random) short- and long-range antiferromagnetic interactions of the special forms between the impurities themselves, see e.g. [12,23]. However, these interactions do not affect the behavior of our disordered correlated spin system qualitatively, compared to the case without direct interaction between impurities. Here, a probability distribution $P(\theta)$ with asymptotics $|\theta|^{-\alpha}$ for large θ leads to a divergence $\chi(T) \propto T^{-1/\alpha}$ which is weaker than that observed above. It was suggested in [9] that the inclusion of other kinds of impurity-impurity couplings (of RKKY form, which violate the exact integrabilty) also do not change the behavior qualitatively. However ferromagnetic impurity-impurity couplings can change the situation drastically, e.g. providing infinite $\chi(T \to 0)$ even for weak disorder. The generalization of our results to correlated electron systems with random impurities will be reported elsewhere.

To conclude, we have constructed exactly the thermodynamics of the Heisenberg antiferromagnetic spin-$\frac{1}{2}$ chain with embedded disordered impurities. The results are of high (numerical) accuracy and valid for arbitrary ranges of magnetic field and temperature. For strong disorder of the impurity-host couplings the local moments are non-quenched. For weak disorder of the host-impurity couplings, on the other hand, spin excitations of the host screen the impurities, but with the non-Fermi-liquid behaviors of the thermodynamic characteristics. The comparison of our theory with the data of a perturbative analysis and with those of magnetic experiments on disordered non-Fermi-liquids in rare-earth alloys shows qualitative agreement.

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